

Experimental corroboration of the Mulheran-Blackman explanation of the scale invariance in thin film growth: the case of InAs quantum dots on GaAs(001).

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Abstract

Mulheran and Blackman have provided a simple and clear explanation of the scale invariance of the island size distribution at the early stage of film growth [Phil. Mag. Lett. **72**, 55 (1995)]. Their theory is centered on the concept of capture zone properly identified by Voronoi cell. Here we substantiate experimentally their theory by studying the scale invariance of InAs quantum dots (QDs) forming on GaAs(001) substrate. In particular, we show that the volume distributions of QDs well overlap the corresponding experimental distributions of the Voronoi-cell areas. The interplay between the experimental data and the numerical simulations allowed us to determine the spatial correlation length among QDs.

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The film growth governed by nucleation and growth is characterized by the emergence of the scale invariance for some relevant quantities linked to the islands forming the film, namely the size and the radial distribution function. In this article we will place emphasis on the former.

Thanks to thorough numerical simulations of two dimensional (2D) thin film growth for very large $R = D/Fa^4$ ratio (where D is the diffusion coefficient of the adatoms, F is the flux of impinging atoms and a is the lattice constant), it was inductively established the following relation for the size (s) distribution of islands in the low coverage (Θ) regime [1][2][3]

$$N_s(\Theta) = \frac{\Theta}{\langle s \rangle^2} f\left(\frac{s}{\langle s \rangle}\right), \quad (1)$$

where $\langle s \rangle$ is the average island size and $f(x)$ is the scaling function. The first experimental evidence of scale invariance and in turn the corroboration of eqn.1, was given by Strosio and Pierce who studied the initial stages of the 2D growth of Fe on Fe(001)[4]. Subsequently, other confirmations came from the study of the strained heteroepitaxial growth of InAs on GaAs(001) in the submonolayer regime [5][6]. However, InAs on GaAs(001) grows in a Stranski-Krastanov mode, so there exists a critical coverage after which the initial epitaxial (2D) growth is followed by the formation of 3D coherent islands or quantum dots (QDs). It is a rather substantiated fact that also the size (i.e. the volume or the number of atoms making up the dot) distribution of QDs satisfies eqn.1 [7][8][9].

At least as far as its derivation is concerned, eqn.1 is quite an empirical relation, often referred to as scaling ansatz, and as we have seen, both experimental data and numerical simulations seem to support it. It owes to Mulheran and Blackman (MB) [10][11] if eqn.1 is, in fact, much more than a simple phenomenological equation, in that they gave a convincing,

neat explanation of its geometric/physical origin. Their argument is rather simple and centers on the concept of capture zone.

To begin with let us consider N_0 points per unit area distributed throughout a plane. To these it is possible to associate a Voronoi network [12] that has the property to tessellate the plane; to each point is associated a cell or polygon. The cells do not overlap one another and the space belonging to the i -th cell is that closer to the i -th point than to any other of the remaining points. This geometric description finds an obvious physical counterpart in film growth: surface, nucleation centers and capture zones in place of plane, points and Voronoi cells, respectively. This analogy, first proposed by Venables and Bell, [13] is based on the conjecture that the adatoms which land in the i -th cell are captured, on average, by the i -th nucleation center, because, by definition, it is the closest one.

It is then easy to convince oneself that, if all the nucleation centers becomes active simultaneously, the size (volume) distribution of islands will resemble the size distribution of the areas of the Voronoi cells because, on average, a particular island is made up by the atoms previously contained in the corresponding Voronoi cell. Incidentally, this conclusion is apparently independent of the dimensionality of the islands. It follows that, since the number of islands (nucleation centers) does not vary with Θ , provided the change of the shape of the Voronoi cells due to the growth of islands [14] is negligible, the size distribution of islands does not change its shape for all the coverage for which islands can be well approximated by dimensionless points. We got to the point: *the scale invariance is nothing but the invariance of the Voronoi network*. As such, even in the simplest case of simultaneous heterogeneous nucleation, discussed above, the scale invariance, in principle, is not an exact law because it rests on the structureless island approximation. In the event, however, this approximation is rather good at small coverage which is, after all, the regime where the scaling behavior is

verified.[1][2][3]

What about non-simultaneous nucleation?

Let us take into account, for the sake of simplicity, the homogeneous nucleation case where dimer is the stable island. Taking it that the growth is negligible during the initial nucleation stage, the rate equations of the process reads

$$\begin{cases} \dot{n}_1 \cong 1 - Rn_1^2 \\ \dot{n} = 2Rn_1^2, \end{cases} \quad (2)$$

where n_1 and n are the number of monomers and stable islands per site, respectively. Since usually $Rn_1^2 \ll 1$, [2] it can be neglected from the first equation in the system eqn.2, thus one ends up with

$$\dot{n} \approx 2R\Theta^2. \quad (3)$$

Eqn.3 points out that a large value of the parameter R entails a fast nucleation. In other words, a large R would involve so a fast nucleation that it could be completed, for all practical purpose, before the growth could become considerable. Such a scenario strongly resemble a simultaneous nucleation event and consequently is a warning sign of scale invariance. As a matter of fact, this is another condition for the island size distribution function to display scale invariance in numerical simulations.[1][2][3]

In conclusion, to the extent that the islands can be considered structureless and as long as the nucleation rate is adequately fast, the associated Voronoi network is quasi-invariant and, as a consequence, quasi-scale invariance follows.

In this letter we display experimental Atomic Force Microscopy (VEECO-Digital) data which support the aforementioned conclusion. In particular, we studied the scale invariance

properties of InAs QDs on GaAs(001).

The details of the experimental set up are reported elsewhere.[9][15] Here it is enough to remark that the entire kinetics was determined by a single shot of InAs on the same GaAs substrate. Our procedure allows us to obtain a rather fine sampling of the range of coverage ($\Delta\Theta = 0.01$ ML) and to avoid the use of a different substrate for each value of the coverage. The substrate was held at 500 °C and the InAs flux was $F = 0.029$ ML/s.

In what it follows we focus only on the so called large QDs. Small QDs are immaterial for what we are going to draw here, because their number soon becomes negligible. [9][15] The nucleation and its rate are shown in fig.1. Their behavior points out that the condition for a possible scale invariance is fulfilled, because the nucleation terminates, in fact, within a time interval much shorter than that necessary to cover the entire surface. In point of fact, the scaled distributions of the QD volumes collapse in a single curve as shown in fig.2a. What is more, the satisfactory agreement between the distribution of the QD volumes and the corresponding distribution of the Voronoi-cell areas displayed in figs.2b-e corroborates MB's theory.

There is still one last subject to be broached: the functional form of the scaling function. Until today there exist only empirical proposals [1] [3] [10] [11], so it is a question of deciding what is the most satisfactory among them, and not only on the basis of a mere numerical utilitarian employ but also, we would say, *above all*, on the epistemological point of view. There can be no doubt that MB's proposal is the preferable one for the simple reason that it is direct consequence of the explanation of the distribution scale invariance. *The distribution function must be the same as that of the Voronoi cells.* This problem has been studying for many years [12] and basically it remains on the conjecture of Kiang [16] that, after having solved the problem in 1D, extended his result (but this is a conjecture) to the 2D case.

Following Kiang, MB proposed the following scale function

$$f_{\beta}(x) = \frac{\beta^{\beta}}{\Gamma(\beta)} x^{\beta-1} e^{-\beta x}, \quad (4)$$

where $\beta \in \mathbb{R}$ is a parameter and $\Gamma(x)$ is the Euler's gamma function.

In ref.[11] this choice is discussed in comparison to the function proposed in ref.[3] where, in turn, the comparison with the proposal of ref.[1] is performed. We have fitted the experimental volume distributions both to eqn.4 and to the distribution of ref.[3], here reported for the sake of clearness

$$f_i(x) = C_i x^i e^{-ia_i x^{1/a_i}}, \quad (5)$$

where i is the number of atoms making up the critical nucleus, C_i and a_i are constants). Although both functions return more than acceptable fits, the χ^2 test is in favor of eqn.4 in four cases out of six. In fig.3 we have plotted the values of β returned by fits as a function of Θ . As it appears β is constant within errors in observance of the scaling law. The average $\bar{\beta} \cong 4.5$ indicates that there exists a certain degree of spatial correlation among the QDs, because poissonian Voronoi tessellation returns $\beta \sim 3.5$. [10] In order to determine the correlation length we performed a series of computer simulations. The substrate is represented by a (3000×3000) square lattice, where we select 720 points ($N_0 = 8 \times 10^{-5}$ very close to the experimental value) according to the rule that two points cannot lay closer than a distance ξ (hardcore correlation approximation [17]). Afterwards the Voronoi tessellation is generated and its distribution function is fitted to eqn.4. Similar as the mean experimental β is obtained for $\xi \cong 30 - 35$ which corresponds to 30 - 35 nm in the real surface. In fig.4 two typical simulations together with their fits are shown.

Finally, a remark is in order. In our previous article [9] we analyzed the scaling properties on the basis of eqn.5, obtaining, on the whole, very good results. As we have discussed above, this is not surprising because often the scattering of experimental data makes it hard to distinguish the slight differences between eqn.4 and eqn.5. The point is that according to eqn.5, we proposed a possible scenario which, in the light of the MB scaling explanation, cannot be entirely correct. In actual fact, we were led astray by the importance assumed by the parameter i in eqn.5, according to which a bell shape distribution function cannot be compatible with $i = 0$. As a consequence, we were led to postulate the existence of a possible "*artifact of the erosion process*" [9] which is, in fact, an unnecessary requirement in the contest of MB's theory. What can be maintained with a high degree of confidence is that the nucleation takes place at the step edges, the step erosion brings about an explosive nucleation and the QD formation proceeds by diffusion and aggregation only[see fig.2]. It remains to expound the exponential distribution disclosed by the small QDs [9], on that we are working on.

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Figure Captions

Fig. 1 - Experimental nucleation function of InAs/GaAs(001) quantum dots (dotted curve) and the nucleation rate (solid curve).

Fig. 2 - Scaled distributions of the experimental island volume for QDs in the range 1.60 – 2.04 ML of InAs coverages (a). Comparison between the distribution of the QD volumes and the corresponding distribution of the Voronoi-cell areas for the following InAs coverages: 1.65 ML (b), 1.68 ML (c), 1.70 ML (d), 1.79 ML (e).

Fig. 3 - Behavior of the parameter β as a function of coverage as obtained by fitting the experimental distributions of Voronoi polygon areas.

Fig. 4 - Typical distributions of simulated Voronoi polygon areas. The simulations have been carried out on a 3000×3000 square lattice with a density $N_0 = 8 \times 10^{-5}$ point-like correlated dots. Correlation lengths $\xi = 30$ (a) and $\xi = 35$ (b) have been used. Data have been fitted to eqn.4 (see the text).

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